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(54) ROOM TEMPERATURE SETTING COMPOSITION

(57)Abstract:
PROBLEM TO BE SOLVED: To obtain a room temperature setting composition having a low viscosity, excellent in handleability, mechanical strength and adhesiveness, and enabling the cure rate to be adjusted in a wide range by making the composition include two kinds of specific polyoxyalkylene polymers and an epoxy resin as essential ingredients.
SOLUTION: This composition comprises (A) a polyoxyalkylene polymer having at least one hydrolyzable silicon group of the formula SiX2R1 [R1 is a 1-20C (substituted)monomvalent organic group; X is OH or the like], (B) a polyoxyalkylene polymer having at least one hydrolyzable silicon group of the formula SiX3 which can be obtained by introducing an unsaturated group into the end of a polyoxyalkylene polymer having at least one OH and then reacting with the mercapto group of a silicon compound (e.g. 3- mercaptopropyltrimethoxysilane or the like) of the formula HS-R2-SiX3 (R2 is a 1-17C divalent hydrocarbon) and (C) an epoxy resin (e.g. a bisphenol A-type epoxy resin or the like).

CLAIMS

[Claim(s)]
[Claim 1]A polyoxyalkylene polymer (A) which has a hydrolytic silicon group expressed with a following formula (1). And after introducing an unsaturation group into an end of a polyoxyalkylene polymer which has a hydroxyl group, A polyoxyalkylene polymer (B) which has a hydrolytic silicon group which is obtained by making a sulfhydryl group of a silicon compound expressed with this unsaturation group and a formula (3) react, and which is expressed with a following formula (2), and a room-temperature-curing natura constituent (D) which uses an epoxy resin (C) as an essential ingradient.

$-SiX_2R^1$ ... (1)

(Among a formula (1), as for substitution of the carbon numbers 1-20 or an unsubstituted univalent organic group, and X, a hydroxyl group or a hydrolytic basis, however two X may be the same, or R^1 may differ.)

$-SiX_3$ ... (2)

(X are the same as the above among a formula (2).) three X may be the same or may differ.

$HS-R^2-SiX_3$ ... (3)

(X are the same as the above among a formula (3).) three X may be the same or may differ. R^2 is a divalent hydrocarbon group of the carbon numbers 1-17.
[Claim 2]The room-temperature-curing nature constituent (D) containing an epoxy curing agent (E) according to claim 1.
[Claim 3]The room-temperature-curing nature constituent (D) containing a compound (F) which has simultaneously a hydrolytic silicon group and reactive functional groups other than hydrolysis nature in the same molecule according to claim 1 or 2.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention]This invention relates to the room-temperature-curing nature constituent hardened under hygroscopic-surface-moisture existence.
[0002]
[Description of the Prior Art]The polyoxyalkylene polymer which has a hydrolytic silicon group at the end is used for the use of a coating composition, seal constituents, etc., such as sealant and adhesives, taking advantage of the feature that a hardened material has rubber elasticity. The polymer which has a hydrolytic silicon group which two hydrolytic bases per silicon atom combine is indicated to JP,61-18582,B, JP,3-72527,A, and JP,3-47825,A as such a polymer combine is indicated. Such a polymer is dramatically excellent in the balance of physical properties, such as being able to obtain a flexible hardened material and excelling also in storage stability.
From performance in use being high, industrial production was actually carried out and the commercial scene is established in the use of adhesives, a water blocking material, etc.
However, although a cure rate can be made to some extent quick by selection of a curing catalyst etc. by one side in the polymer which has a hydrolytic silicon group which two hydrolytic bases per silicon atom combine, Making it improve by leaps and bounds had a limit from reactivity with the water of a silicon group, and the limit was among the uses asked for a quick cure rate naturally.
[0003]moreover -- it is a polymer which has a hydrolytic silicon group which three hydrolytic bases per silicon atom combine with JP,58-10418,B and JP,58-10430,B -- a molecular weight -- 6000 or less -- the hardenability polymer of low molecular weight is indicated comparatively. Since reactivity with water is high, the hardenability polymer which has a hydrolytic silicon group which three hydrolytic bases per such a silicon atom combine has the characteristic that a cure rate is very quick, but. The molecular weight was inferior to the elongation and pliability of the hardened material which are acquired from 6000 or less and it being low molecular weight comparatively by hardening.
[0004]As a method of improving storage stability in the polymer which has a hydrolytic silicon group which three hydrolytic bases per silicon atom combine, The polymer obtained by the urethane-ized reaction of polyoxypropylene polyol and an isocyanate substitution type trialkoxysilane compound in JP,10-245482,A, The constituent which consists of a curing catalyst and amino group substitution alkoxysilane, Or in JP,10-245484,A, the mixture of the polyoxyalkylene polymer whose polyoxyalkylene polymer and molecular terminal whose molecular terminal is the Tori alkoxy silyl groups are an alkyl dialkoxy group is indicated. The polymer intrinsically obtained by the urethane-ized reaction of polyoxypropylene polyol and an isocyanate substitution type trialkoxysilane compound in this method is used. In order the reactivity of the polyoxypropylene polyol and the isocyanate group in this method is low and to obtain significant reaction velocity, Although use of reaction accelerators, such as various kinds of metal salt which a urethane-ized reaction may be sufficient as and is known for promotion of a reaction, was desirable, there was a case where alkoxy silyl groups caused hydrolysis and crosslinking reaction, and a polymer served as hyperviscosity with a small amount of moisture which remains in the system of reaction in that case, plentifully. Since it became hyperviscosity even if it originated in the urethane bond itself, it had a problem in workability.

[0005]By one side, it could not say that the polyoxyalkylene polymer which has a hydrolytic silicon group at the above-mentioned end was never excellent about the intensity of a hardened material,

and adhesive strength with adherend, but the use was restricted naturally. As a method of improving an adhesive property, the method of using together the polymer which has a hydrolytic silicon group, and an epoxy resin is proposed by JP,61-268720,A and JP,7-2828,B. Although it was effective in such a method improving the intensity and adhesive strength of a hardened material, in respect of the cure rate, it was not yet enough, and when using it especially as adhesives, the adhesive property was wanted to be revealed in shortest possible time.
[0006]And it applies for the room-temperature-curing nature constituent which uses as an essential ingredient the polymer and epoxy resin which have a hydrolytic silicon group which three hydrolytic bases per silicon atom combine in ten to Japanese-Patent-Application-No. 204041 specification by these people. As a polyoxyalkylene polymer which has a hydrolytic silicon group which three hydrolytic bases per silicon atom combine in this method, Although the polymer obtained by making the trimethoxysilane which is a hydrosilyl compound react to the end of a raw material polyoxyalkylene polymer to the polymer which introduced the unsaturation group is used, The trimethoxysilane which is the hydrosilyl compound used here had big difficulty on [, such as a transfer and storage,] handling art from the problem of safe according to the disproportionation to Silang (SiH_4).
[0007]In this method, when hydrolysis and crosslinking reaction of a hydrolytic silicon group advanced promptly, while the cure rate of resin became quick, available time and what is called an open time had the fault [that it is short] of having elapsed and being hard to use it. Adjustment of adjusting freely generally, so that adjustment of the cure rate did not receive influence in the reactivity of the end although the thing possible to some extent was known by the kind of the various metal carboxylate which has a catalysis of a hardening reaction, basicity, an acidic compound, etc., etc., or selection of quantity It is called for that it can respond to various cure rates also industrially, and controllable art was freely searched for about hardenability.
[0008]
[Problem(s) to be Solved by the Invention]In view of the above-mentioned problem, the polymer which has a hydrolytic silicon group industrially obtained easily considering the silane compound [handling / a silane compound] as a raw material is used for the purpose of this invention, It is dealt with by hypoviscosity, and excels in a sex, and excels in the mechanical strength of the hardened material after hardening, and adhesive strength with adherend, and is in moreover providing the hardenability constituent which can be adjusted in the range with a wide cure rate.
[0009]
[Means for Solving the Problem]Namely, a polyoxyalkylene polymer (A) in which this invention has a hydrolytic silicon group expressed with a following formula (1), And after introducing an unsaturation group into an end of a polyoxyalkylene polymer which has a hydroxyl group, . Ara obtained by making a sulfhydryl group of a silicon compound expressed with this unsaturation group and a formula (3) react, A polyoxyalkylene polymer (B) which has a hydrolytic silicon group expressed with a following formula (2), and a room-temperature-curing nature constituent (D) which uses an epoxy resin (C) as an essential ingredient are provided.

[0010]- SiX_2R^1 ... (1)

(Among a formula (1), as for substitution of the carbon numbers 1-20 or an unsubstituted univalent organic group, and X, a hydroxyl group or a hydrolytic basis, however two X may be the same, or R^1 may differ.)

- SiX_3 ... (2)

(X are the same as the above among a formula (2).) three X may be the same or may differ.

$HS-R^2-SiX_3$ ... (3)

(X are the same as the above among a formula (3).) three X may be the same or may differ. R^2 is a divalent hydrocarbon group of the carbon numbers 1-17.
[0011]
[Embodiment of the Invention](Polyoxyalkylene polymer) The polyoxyalkylene polymer (B) which has a hydrolytic silicon group expressed with the polyoxyalkylene polymer (A) and the above-mentioned formula (2) which have a hydrolytic silicon group expressed with the above-mentioned formula (1) used in this invention so that it may state below, It is preferred to usa as a raw material the

polyoxyalkylene polymer which has a functional group, to introduce a hydrolytic silyl group into a part or all of the functional group via an organic group, and to be manufactured. For example, what is indicated to JP,3-47825,A, JP,3-72527,A and JP,3-79627,A is mentioned.

(Raw material polyoxyalkylene polymer) As the polyoxyalkylene polymer (A) used in this invention, and a raw material polyoxyalkylene polymer of (B), the thing of the hydroxyl group end which makes cyclic ether etc. react and is manufactured is preferred under existence of a catalyst and existance of an initiator. This raw material polyoxyalkylene polymer may be on a straight chain, may be a letter of branching, or there may be with these mixtures.

[0012]As an initiator, the hydroxy compound etc. which have one or more hydroxyl groups can be used. As cyclic ether, ethylene oxide, propylene oxide, butylene oxide, hexylenoxide, a tetrahydrofuran, etc. are mentioned. As a catalyst, alkaline metal catalysts, such as a potassium system compound and a caesium system compound, a composite metal cyanide complex catalyst, a metalloporphyrin catalyst, etc. are mentioned.

[0013]As a functional group number, the raw material polyoxyalkylene polymer of 2-8 is preferred, and when a functional group number uses the thing of 2 or 3 as a raw material, it is especially desirable from the polyoxyalkylene polymer (A) and (B) which demonstrates the adhesive strength which was excellent greatly in pliability as the hardened material characteristic being obtained. as a raw material polyoxyalkylene polymer, the thing of the amount of polymers is preferred — further — the ratio of weight average molecular weight (Mw) and a number average molecular weight (Mn) — the narrow thing of molecular weight distribution with small Mw/Mn is preferred.

[0014]When the raw material polyoxyalkylene polymer of Mw/Mn which is different with the same number average molecular weight (Mn) compares, it has the feature that the viscosity of a polymer becomes low and the thing which has small Mw/Mn is excellent in workability. When it is going to adjust the viscosity of a polymer to a certain fixed level, the content of the polymer of low molecular weight decreases as the small thing of Mw/Mn. The polyoxyalkylene polymer which has by this a hydrolytic silicon group obtained considering this as a raw material, As for the hardened material produced by excelling in the hardening characteristics — the depths hardenability at the time of hardening becomes good — compared with the case where the large thing of Mw/Mn is used as a raw material, and hardening, the elongation of a hardened material serves as high intensity large also what has a the same elastic modulus.

[0015]The raw material polyoxyalkylene polymer of the amount of polymers, It can obtain by the method of using a composite metal cyanide complex catalyst, the method of quantifying many and carrying out polymers quantification by [which manufactured using the alkali catalyst etc.] making many halogenated compounds, such as a methylene chloride, the polyoxyalkylene polymer of low molecular weight react comparatively, etc. As for the small raw material polyoxyalkylene polymer of Mw/Mn, what is obtained considering a composite metal cyanide complex as a catalyst is preferred. The complex which uses zinchexacyano cobaltate as the main ingredients as a composite metal cyanide complex is preferred, and ether and/or an alcoholic complex are especially preferred. The presentation can use what is intrinsically indicated to JP,46-27250,B. In this case, as ether, ethyleneglycol dimethyl ether (glyme), diethylene glycol dimethyl ether (jig lime), etc. are preferred, and especially glyme is preferred from a point of the handling at the time of manufacture of a complex. As alcohol, t-butanol is preferred.

[0016]As a molecular weight of a raw material polyoxyalkylene polymer. the thing of 6,000-50,000 is preferred at a number average molecular weight (Mn), and, specifically, especially the thing of 8,000-20,000 is preferred. As Mw/Mn of a raw material polyoxyalkylene polymer, 1.7 or less are preferred, 1.6 or less are still more preferred, and 1.5 especially or less are preferred.

[0017]As a raw material polyoxyalkylene polymer, the copolymer of a polyoxyethylene, polyoxypropylene, polyoxy butylene, polyoxy hexylene, polyoxy tetramethylen, and two or more sorts of cyclic ether is specifically mentioned. Especially a desirable raw material polyoxyalkylene polymer is the polyoxypropylene polyol of 2 - 6 value, and are polyoxypropylene diol and polyoxypropylene triol especially. When using for the method of the following (b) or (**), the polyoxyalkylene polymer of unsaturation groups, such as an allyl end polyoxypropylene monooar, can also be used.

(Polyoxyalkylene polymer (A)) A polyoxyalkylene polymer (A) has a hydrolytic silicon group expressed with the end or side chain of a chain with a following formula (1).

[0018]- SiX_2R^1 ... (1)

(Among a formula (1), as for the substitution of the carbon numbers 1-20 or an unsubstituted univalent organic group, and X, a hydroxyl group or a hydrolytic basis, however two X may ba tha same, or R^1 may differ.)

R^1 in formula (1) is a univalent organic group the substitution of the carbon numbers 1-20, or unsubstituted, and a with a carbon number of eight or less alkyl group, a phenyl group, or a fluoro alkyl group is preferred. A methyl group, an ethyl group, a propyl group, a butyl group, a hexyl group, a cyclohexyl group, a phenyl group, etc. are raised, and, specifically, especially a methyl group is preferred. when two or more R^1 exist, those R^1 may be the same, or may differ.

[0019]As a hydrolytic basis in X, a halogen atom, an alkoxy group, an acyloxy group, an alkenyloxy group, a carbamoyl group, an amino group, an aminooxy group, a KETOKISHI mate group, etc. are mentioned, for example. As for the carbon number of the hydrolytic basis which has a carbon atom among these, six or less are preferred, and four especially or less are preferred. The viewpoint of hydrolysis nature being quiet and being easy to deal with it among these to especially an alkoxy group is preferred. Especially as an alkoxy group, a methoxy group and an ethoxy basis are preferred, and a methoxy group is the most preferred.

[0020]That is, it is preferred that it is especially the structure of having an alkyl dialkoxy silyl group, as a hydrolytic silicon group expressed with a following formula (1), and a methyl dimethoxy silyl group is the most preferred. The hydrolytic silicon group expressed with a formula (1) is usually introduced into a raw material polyoxyalkylene polymer via an organic group. That is, as for a polyoxyalkylene polymer (A), it is preferred to have a basis expressed with a formula (4).

[0021]-R^6-SiX_2R^1 ... (4)

(A divalent organic group, R^1, and X of R^6 are the same as that of the above among a formula (4).) Although the method in particular of introducing a hydrolytic silicon group to a raw material polyoxyalkylene polymer is not limited, it can be introduced, for example by (b) of the following - the method of (**).

(**) A method to which the hydrosilyl compound expressed with it by a formula (5) after introducing an unsaturation group into the end of the polyoxyalkylene polymer which has a hydroxyl group is made to react.

[0022]$HSiX_2R^1$ ... (5)

(R^1 and X are the same as the above among a formula (5).)

The method of making the compound which has an unsaturation group and a functional group react to the terminal hydroxyl groups of the polyoxyalkylene polymer which has a hydroxyl group, and combining it by the ether bond, the ester bond, a urethane bond, or carbonate combination as a method of introducing an unsaturation group, is mentioned. As an unsaturation group here, CH_2=CH-R'- (R' is a divalent hydrocarbon group of the carbon numbers 1-18) is preferred. As for R', it is preferred that it is a hydrocarbon group of the carbon numbers 1-5. Especially as an unsaturation group, an allyl group is preferred. Especially as a compound which has an unsaturation group and a funotional group, allylchloride is preferred, and the hydroxyl group of the polyoxyalkylene polymer which has a hydroxyl group can be used as an allyloxy group by using this. When polymerizing alkylene oxide, the method of introducing an unsaturation group into the side chain of a raw material polyoxyalkylene polymer can also be used by adding and carrying out copolymerization of the unsaturation group content epoxy compounds, such as allyl glycidyl ether.

[0023]When making a hydrosilyl compound react, catalysts, such as a platinum system catalyst, a rhodium system catalyst, a cobalt system catalyst, a palladium system catalyst, and a nickel series catalyst, can be used. Platinum system catalysts, such as chloroplatinic acid, platinum metal, a platinum chloride, and a platinum olefin complex, are preferred. As for the reaction to which a hydrosilyl compound is made to react, it is preferably preferred to carry out at the temperature of 50-120 ** for several hours 30-150 **.

[0024](**) A method to which the compound expressed with the end of the polyoxyalkylene polymer which has a hydroxyl group by a formula (6) is made to react.

R^1-SiX_2-R^2NCO ... (6)

(R^1 and X are the same as the above among a formula (6).) R^2 is a divalent hydrocarbon group of the

carbon numbers 1-17.
A publicly known urethane-ized catalyat may be used in the case of tha above-mentioned reaction. As for the above-mentioned reaction, it is preferably preferred to oarry out at the temperature of 50-150 ** for several hours 20-200 **.
[0025](**) After making polyisocyanate oompounds, such aa tolylane diisooyanate, react to the and of the polyoxyalkylene polymer which has a hydroxyl group and oonsidering it as an iaocyanate group end, Method $R^1-SiX_2-R^2W$ to which W basis of the silicon compound expressed with a formula (7) to this isocyanate group is made to react ... (7)
(R^1, R^2, and X are the same as the above among a formula (7).) Active hydrogen containing group as which W is chosen from a hydroxyl group, a carboxyl group, a sulfhydryl group, and an amino group (the 1st class or the 2nd class).
(**) A method to which the unsaturation group and the sulfhydryl group of a silicon compound expressed with the formula (7) whose W is a sulfhydryl group are made to react after introducing an unsaturation group into the end of the polyoxyalkylene polymer which has a hydroxyl group.
[0026]The method and unsaturation group which introduce an unsaturation group are the same as that of what was explained in (b). 3-mercapto propylmethyl dimethoxysilane etc. are mentioned as a silicon compound by which W is expressad with the formula (7) which is a sulfhydryl group.
Polymerization initiators, such aa a radical generator, may be usad, and it may be made to react with radiation or heat in the caae of the above-mentioned reaction, without using a polymerization initiator depending on the case. As a polymerization initiator, a polymerization initiator, a metal compound catalyat, etc. of a peroxide system, azo, or a redox system are mantioned, for example. Specifically aa a polymerization initiator, 2,2'-azobisisobutyronitrile, 2,2'-azobis 2-methylbutyronitrila, benzoyl peroxide, t-alkyl peroxy ester, acetyl peroxide, diisopropyl peroxy carbonate, eto. are mentioned. As for the above-mentioned reaction, it is preferably preferred to carry out at 50-150 ** for several hours - tens hours 20-200 **.
(Polyoxyalkylene polymer (B)) A polyoxyalkylene polymer (B) has a hydrolytic silicon group expressed with the end or side chain of a chain with a following formula (2).
[0027]$-SiX_3$ ... (2)
(X are the same as the above among a formula (2).) three X may be the same or may differ.
Since reactivity is high, the polymer which has a hydrolytic silicon group expressed with the above-mentioned formula (2) has the characteristic that the cure rate at the time of hardening is very quick. As a hydrolytic basis in X in a formula (2), As well as the above, for example, a halogen atom, an alkoxy group, an acyloxy group, An alkenyloxy group, a carbamoyl group, an amino group, an aminooxy group, a KETOKISHI mate group, a hydride group, etc. are mentioned, an alkoxy group is preferred in these, a methoxy group and an ethoxy baaia are specifically preferrad, and a methoxy group ia tha moat preferred.
[0028]That is, it is preferrad that it is especially the atructure of having the Tori alkoxy silyl groups, as a hydrolytic ailicon group expreased with a following formula (2). A trimethoxysilyl group is the moat preferred. As for a polymer (B), it is preferred that the hydrolytio silicon group expressed with a formula (2) is introduced into a raw material polyoxyalkylene polymer via an organic group like a polymer (A). That is, as for a polyoxyalkylene polymer (B), it is preferred to have a basis expressed with a formula (8).
[0029]$-Y-S-R^2-SiX_3$ ... (8)
(X are the same as the above among a formula (8).) three X may be the same or may differ. Y is a divalent hydrocarbon group of 1-17. R^2 is a divalent hydrocarbon group of the carbon numbers 1-17. As Y, the hydrocarbon group of the carbon numbers 1-5 is preferred, and especially a trimethylene group is preferred. As R^2, the hydrocarbon group of the carbon numbers 1-5 is preferred, and especially a trimethylene group is preferred.
[0030]A polyoxyalkylene polymer (B) is obtained by making the sulfhydryl group of a silicon compound expressed with this unsaturation group and a formula (3) reaot, after introducing an unsaturation group into the end of the polyoxyalkylena polymer which has a hydroxyl group.
$HS-R^2-SiX_3$ ... (3)

(X are the same as the above among a formula (3).) three X may be the same or may differ. R^2 is a divalent hydrocarbon group of the carbon numbers 1-17.
About the details of the introducing method, it is the same with having explainad in method (**) which introduces the hydrolytic silicon group of said polyoxyalkylene polymer (A). Aa an unsaturation group, $CH_2=CH-R'-$ (R' is the same as the above) is preferred as above-mentioned.
[0031]As a silicon compound expressed with a formula (3), 3-mercapto propyltrimethoxysilane, 3-mercaptopropyl triethoxysilane, etc. are mentioned. It has the characteristic that the cure rate of the polyoxyalkylene polymer obtained with a described method is large, and has the feature of excelling in workability by hypoviscosity. Acquisition of a raw material is also easy and industrially useful.
(Epoxy resin (C)) As an epoxy resin (C) used for this invention, A publicly known thing can be used widely conventionally and it is in a concrete target. Bisphenol A type epoxy resin, Fire retardancy type epoxy resins, such as bisphenol F type epoxy resin and a glycidyl ether type epoxy resin of tetrabromobisphenol A, Cresol novolak type epoxy resin, bisphenol A novolak type epoxy resin, Novolak type epoxy resin, such as bisphenol F novolak type epoxy resin, A hydrogenation bisphenol A type epoxy resin, the glycidyl ether type epoxy resin of bisphenol A / propylene oxide addition, 4-glycidyloxy benzoic acid glycidyl, phthalic acid diglycidyl, Diglycidyl ester system epoxy resins, such as tetrahydrophtal acid diglycidyl and hexahydrophthalic acid diglycidyl, m-aminophenol series epoxy resin, a diaminodiphanylmethane system epoxy resin, Glycidyl ester-typed-epoxy-reain, glycidyl amine-type-epoxy-resin, isocyanurate type epoxy resin, urethane modifiad epoxy reain, various cycloaliphatic-epoxy-reain, and N,N-diglyoidyl aniline, N,N-diglycidyl o-toluidine, Although the epoxidation thing of unsaturation polymars, such aa glycidyl ether of polyhydric alcohol, auch as triglycidyl isocyanurate, polyalkylene glycol diglycidyl ether, and glycerin, a hydantoin type epoxy resin, a resorcinol type epoxy resin, and petroleum resin, etc. are raised, It is not limited to these. A bisphenol A type epoxy resin, bisphenol F type epoxy resin, novolak type epoxy resin, and especially the epoxy resin of a phthalic acid diglycidyl ester system are [among these] preferred.
[0032]As an epoxy resin (C), what contains two or more epoxy groups in a molecule has high reactivity when hardening, and a hardened material is especially preferred from points — it is easy to build the three-dimensional network structure.
(Epoxy curing agent (E)) As for the room-temperature-curing nature constituent (D) of this invention, it is still more preferred to contain an epoxy curing agent (E). As an epoxy curing agent (E), conventionally, can use a publicly known thing widely and specifically Diethylenetriamine, Triethylenetetramine, tetraethylenepentamine, diethylamino propylamine, N-aminoethyl piperazine, m-xylylene diamine, m-phenylenediamine, Diaminodiphenylmethane, diaminodiphenyl sulfone, isophoronediamine, Amines or those salts, such as 2,4,6-tris(dimethyl aminomethyl) phanol, Aldimine, enaminea, polyamide resin, imidazola derivatives, and dicyandiamidas. Boron trifluoride complex compounds, phthalic anhydride, a hexahydrophthalic anhydrida, A tetrahydrophthalic anhydride and methylene tetrahydro phthalio anhydride, Anhydrous carboxylio acid, such aa a DODESHINIRU succinic anhydride, pyromallitic dianhydride, and anhydrous KUROREN acid. polyalkylene oxide aystem polymers (and amination polyoxypropylene glycol.) which average tha basis which can react to an epoxy group and it has at least one piece in intramolecular, such as phenoxy resin, carboxylic acid, and alcohols An end end carboxylation polyoxypropylene glycol etc. A hydroxyl group, a carboxyl group, Although liquefied end functional group content polymers etc. which were embellished with the amino group etc., such as polybutadiene, hydrogenation polybutadiene, an acrylonitrile butadiene copolymer, and an acrylic polymer, are raised, it is not limited to these. 2,4,6-tris(dimethyl aminomethyl) phenol is [among these] the most preferred.
[0033]A compound like ketimines as shown with the following formula (9) or (10), and silazanes can also be used.
$R^3R^4C=N-R^5-NH-R^6-N=CR^7R^8$ ... (9)
(The inside of a formula, R^3, R^4, R^7, and R^8 are a hydrogen atom, a halogen atom, or a univalent hydrocarbon group.) R^5 and R^6 are divalent hydrocarbon groups.
$R^9R^{10}C=N-R^{11}-N=CR^{12}R^{13}$ ... (10)
(The inside of a formula, R^9, R^{10}, R^{12}, and R^{13} are a hydrogen atom, a halogen atom, or a univalent hydrocarbon group.) R^{11} is a divalent hydrocarbon group.

Since amine was generated only after these compounds reacted to moisture, when it can use it as an insidious hardening agent and is especially used in combination of one component type, etc., as compared with other epoxy curing agents, it has the feature that storage stability is excellent.

[0034]Although the compound etc. which are obtained by the dehydration of carbonyl compounds, such as polyemine, methyl ethyl ketone, etc., such as diethylenetriamine, are raised as a typical example, it is not limited to this. The amino group of the ketimines shown by the above-mentioned formula (9) Styrene oxide. Butyl glycidyl ether, 3-glycidyloxypropyl trimethoxysilane. The compound made to react to epoxy group containing compounds, such as 3-glycidyloxy propylmethyl dimethoxysilane and 3-glycidyloxy propyl triethoxysilane, and mono- isocyanate compounds, such as a phenylisocyanate, can also be used.

(Compound (F) which has simultaneously a hydrolytic silicon group and a reactive functional group besides hydrolytic silicon Motomochi) The room-temperature-curing nature constituent (D) of this invention may contain further the compound (F) which has simultaneously a hydrolytic silicon group and a reactive functional group besides hydrolytic silicon Motomochi. A compound (F) is a compound which has a different functional group of at least two kinds of reactivity in one molecule, and makes a hydrolytic silyl group indispensable, and means the compound usually known as a silane coupling agent or an adhesion grant agent. As a compound (F), a conventionally publicly known compound which is indicated, for example to "the optimal selection and use art of a coupling agent, and an appraisal method (TECHNICAL INFORMATION INSTITUTE)" etc. can be used. When a room-temperature-curing nature constituent (D) contains a compound (F), it is desirable from adhesive strength with adherend acting as Kougami further.

[0035]As a hydrolytic silicon group in a compound (F), the hydrolytic silicon group expressed with a formula (1) and e formula (2) is preferred. As a reactive functional group besides hydrolytic silicon Motomochi in a compound (F), an epoxy group, an emino group, a sulfhydryl group, an acrylexy (meta) group, a carboxyl group, etc. are preferred.

[0036]As a compound (F), epoxy group content Silang, amino group content Silang, sulfhydryl group content Silang, acryloxy (meta) group content Silang, and carboxyl group content Silang are mentioned. As epoxy group content Silang, specifically 3-glycidyloxypropyl trimethoxysilane, 3-glycidyloxy propylmethyl dimethoxysilane, 3-glycidyloxy propyl triethoxysilane, 3-(N,N-diglycidyl) aminopropyl trimethoxysilane, N-glycidyl N,N-bis[3-(methyl dimethoxy silyl) propyl] amine, N-glycidyl N,N-bis[3-(trimethoxysilyl) propyl] amine, etc. are mentioned.

[0037]As amino group content Silang, specifically 3-aminopropyl trimethoxysilane, 3-aminopropyl triethoxysilane, 3-aminopropyl methyl dimethoxysilane, N-(2-aminoethyl)-3-aminopropyl trimethoxysilane, N-(2-aminoethyl)-3-aminopropyl methyl dimethoxysilane, N-(2-aminoethyl)-3-aminopropyl triethoxysilane, N,N-bis[3-(methyl dimethoxy silyl) propyl] amine, N,N-bis[3-(methyl dimethoxy silyl) propyl] ethylenediamine, N,N-bis[3-(trimethoxysilyl) propyl] ethylenediamine, N-[(3-trimethoxysilyl) propyl] diethylenetriamine [$H_2N(C_2H_4NH)_2C_3H_6Si(OCH_3)_3$], N-[(3-trimethoxysilyl) propyl] triethylenetetramine [$H_2N(C_2H_4NH)_3C_3H_6Si(OCH_3)_3$], 3-ureido propyl triethoxysilane, N-(N-vinylbenzyl 2-aminoethyl)-3-aminopropyl trimethoxysilane, 3-anilino propyltrimethoxysilane etc. are mentioned.

[0038]As sulfhydryl group content Silang, 3-mercapto propyltrimethoxysilane, 3-mercaptopropyl triethoxysilane, 3-mercapto propylmethyl dimethoxysilane, 3-mercapto propylmethyl diethoxy SHISHIRAN, etc. are specifically raised. As (methacryloyloxy group content Silang, 3-methacryloyl oxypropyl trimethoxysilane, 3-acryloyloxypropyl trimethoxysilane, 3-methacryloyl oxypropyl methyl dimethoxysilane, etc. are specifically mentioned.

[0039]As carboxyl group content Silang, specifically 2-carboxyethyl triethoxysilane, 2-carboxyethyl phenylbis(2-methoxyethoxy)Silang, N-(N-carboxylmethyl 2-aminoethyl)-3-aminopropyl trimethoxysilane, etc. are mentioned. The reactant produced by making two or more sorts of these compounds react may be used. As an example of a reactant, the reactant of amino group content Silang and epoxy group content Silang, The reactant of epoxy group content Silang and sulfhydryl group content Silang, the reactant of sulfhydryl group content Silang, the reactant of amino group content Silang and (meth)acryloyloxy group content Silang, etc. are mentioned. These reactants are easily obtained by mixing the above-mentioned compound and agitating in a room temperature ~150 ** temperature requirement for 1 to 8 hours.

[0040]A compound (F) may be used alone and may be used together two or more kinds.

(Room-temperature-curing nature constituent (D)) The polyoxyalkylene polymer (A) in the room-temperature-curing nature constituent (D) of this invention and the rate of (B) can be arbitrarily chosen according to a use, the characteristic to need, etc.

[0041]Although the polyoxyalkylene polymer (A) and the rate of (B) are arbitrary, (A):(B) =5 - 95:95 - 5 are preferred at a weight ratio, (A):(B) =20 - 80:80 - 20 are still more preferred, and (A):(B) =30 - 70:70 - 30 are the most preferred. That is, adjustment of hardenability becomes possible in the wide range by adjusting a polyoxyalkylene polymer (A) and the rate of (B) arbitrarily. Hardenability can specifically be made quick, so that the rate of a polyoxyalkylene polymer (B) is enlarged, it is lessening the rate of a polyoxyalkylene polymer (B), and the constituent which has hardenability arbitrary to the thing conventionally near equivalent hardenability can be obtained. When what has a large rate of a polyoxyalkylene polymer (B) is used as adhesives, a sealing material, etc., Since hardenability is quick, the effect that the manifestation of the adhesive intensity after construction is quick is acquired, in the case where he would like to make hardening under low temperature quick etc., it is useful to paste up in the directions for use which must be fixed so that adherend may not be moved, during a short time until an adhesive property is revealed especially. On the other hand, from the thing which has a large rate of a polyoxyalkylene polymer (A) being excellent about the extension characteristic of a hardened material. It is important to obtain the optimal constituent each time by changing a polyoxyalkylene polymer (A) and the rate of (B) arbitrarily according to a use or the characteristic to need.

[0042]Although the rate of the polyoxyalkylene polymer (A) and the epoxy resin (C) to (B) in the room-temperature-curing nature constituent (D) of this invention can be chosen arbitrarily, 1 - 300 weight section is preferred to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B), and especially 5 - 100 weight section is preferred. When less than this, the intensity of the hardened material after hardening becomes insufficient, and an epoxy resin (C) becomes insufficient [ductility] in being more than this, and is not preferred.

[0043]Although the rate of the polyoxyalkylene polymer (A) and the compound (F) to (B) in the room-temperature-curing nature constituent (D) of this invention can be chosen arbitrarily, 0 - 30 weight section is preferred to a total of 100 copies of a polyoxyalkylene polymer (A) and (B), and especially 0.1 - 10 weight section is preferred. When a compound (F) is added across a mentioned range, it is not desirable from properties balance, such as the handling nature of a constituent, being spoiled, and the cost of a constituent becoming high etc.

[0044]Although it is not indispensable, the room-temperature-curing nature constituent (D) of this invention does not need to be included even if the additive agent as shown below is included.

Hereafter, an additive agent is explained.

(Bulking agent) A publicly known bulking agent can be used as a bulking agent. Especially the amount of the bulking agent used has 50 - 250 preferred weight section 0.001 to 1000 weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B). The following are mentioned as an example of a bulking agent. These bulking agents may be used independently and may be used together two or more sorts.

[0045]Heavy calcium carbonate with a mean particle diameter of 1-20 micrometers, precipitated calcium carbonate with a mean particle diameter of 1-3 micrometers manufactured with the sedimentation method, The colloid calcium carbonate which carried out the surface treatment of the surface with fatty acid or a resin acid system organic matter, Calcium carbonate, such as minor nature calcium carbonate, fumes silica, sedimentation nature silica, Surface silicondization silica pulverized coal, a silicic acid anhydride, hydrous silicic acids, and carbon black, Magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, resin beads, wood flour, pulp, a cotton chip, mica, and the blacking wash farina --- rubbing --- powder state bulking agents, such as farina, graphite, aluminum impalpable powder, and the Flint powder. Fibrous fillers, such as asbestos, glass fiber, a glass filament, carbon fiber, the Kevlar textiles, and a polyethylene fiber.

[0046]The publicly known hollow body of minerals or the quality of organicity can be used. Since specific gravity is low, workability of a hollow body improves --- a weight saving and the cobwebbing nature of a constituent are improved in a constituent and its hardened material. As an inorganic hollow body, can illustrate a silicic acid system hollow body and a non-silicic acid system hollow body. and as a silicic acid system hollow body, A mitt balloon, perlite, glass balloons, a silica balloon, and fly

ash balloons, As a non-ailicic acid system hollow body, an alumina balloon, a zirconia balloon, a carbon balloon, etc. can be illustrated, a milt balloon and especially glass balloons are preferred, and glass balloons are the most preferred.

[0047]Although there is no limitation in particular in the mean particle diameter of glass balloons, and average particle density. The mean particle diameter of about 10-500 micrometera preferably Usually, about 30-100 micrometers, The average particle density of about 0.1-0.6g/cc preferably About 0.15-0.3g/cc. about 0.05-0.5g/cc ****** -- desirable -- about 0.07-0.3g [cc] /and a pressure resistance [of 10-1000kg/cm] ^{2} grade — a 15 - 300 kg/cm^2 grade and the thing of not less than 90% of ****** are preferably common. Although there is no limitation in particular in the shape of glass balloons, etc., the thing nearer to a real ball has the more preferred viscosity of a constituent from becoming low.

[0048]As a hollow body of the quality of organicity, the hollow body of thermosetting resin and the hollow body of thermoplastics can be illustrated, As a hollow body of thermosetting resin, a phenol balloon, an epoxy balloon, and a urea balloon as a hollow body of thermoplastics, A saran balloon, a polystyrene balloon, a polymethacrylate balloon, a polyvinyl alcohol balloon, and a styrene acrylic balloon can be illustrated. What coated the surface of the hollow body of thermoplastics with thermosetting resin, and the hollow body of thermoplastics which constructed the bridge can also be used. As particles, the porous amoty capsid which has a hole what is called other than a hollow body is raised, and it may be made to foam. after blending what that to which it foamed beforehand may be sufficient as, and contains a foaming agent.

[0049]When the hollow body of the quality of organicity is used, the hardenability constituent which has the feature of excelling in elongation with a low modulus in the tractive characteristics of the hardened material after hardening besides the effect which carries out the weight saving of a constituent and its hardened material can be obtained. It is raised by the hollow body of the hybrid type which coated the surface of the organic hollow body with inactive inorganic powder, and specifically, What coated the surface of the hollow body which uses polyacrylonitrile as the main ingredients with granular materials, such as calcium carbonate, talc, and titanium oxide, is preferred from the good thing of familiarity by other bulking agents.

[0050]As an example of an inorganic hollow body, as a milt balloon, for example a win light (IJICHI Chemicals) etc., As glass balloons, it is a Scotch whisky light. Glass BABURUZU (3M), CEL-STAR (Tokai industry), Q-CEL (Pacific chemicals), MICRO BALLOON (EMERSON & CUMING), CELAMIC GLASS MODULES (PITTSBURGH CORNING CORP.) etc., As fly ash balloons, CEROSPHERES (PFA MARKETING LTD.), FILLITE (FILLITE U.S.A, INC.) etc., As an alumina balloon, as BW (Showa Denko) and a zirconia balloon, HOLLOW ZIRCONIUMSPHERES (ZIROQA) etc., As a carbon balloon, KUREKASU fair (Kuraha chamicals), car boss fair (GENERAL TECHNOLOGIESCORP.), etc. are raised.

[0051]As an example of the hollow body of the quality of organicity, as a phenol balloon, for example PHENOLIC MICROBALLOONS (UCC) etc., As an epoxy balloon, ECOSCSPHERES EP (EMERSON & CUMING) etc., As a urea balloon, ECOSCSPHERES VF-0 (EMERSON & CUMING) etc., As a saran balloon, SARANMICROSPHERES (DOWCHEMICAL COMPANY), Expancel (Japanese phyllite), the Matsumoto microsphere (Matsumoto Yushi-Seiyaku), etc., As a polystyrene balloon, DYLITE EXPANDABLE POLYSTYRENE (ARCO POLYMERS INC.), EXPANDABLE POLYSTYRENE BEADS (BASF WYANDOTE CORP.) etc. are raised for SX863 (P) (Japan Synthetic Rubber) etc. as a constructed type styrene acrylic acid balloon of a bridge.

[0052]As a hollow body of the hybrid type which coated the surface of the organic hollow body with inactive inorganic powder, Matsumoto microsphere MFL series (Matsumoto Yushi-Seiyaku) etc. can be illustrated. 0.3 - 40 weight section is still more preferably preferred [the amount of the hollow body used] 0.1 to 50 weight section preferably 0.01 to 100 weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B).

[0053]If a point with required taking care that a hollow body does not break according to the shearing force at the time of mixing in mixing of a hollow body, especially the hollow body of comparatively weak construction material like glass balloons is removed, it can treat like the usual bulking agent. (Plasticizer) A publicly known plasticizer can be used as a plasticizar. The amount of the plasticizer used has 0.001 - 1000 preferred weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B). The following are mentioned as an example of a plasticizer.

[0054]Phthalic ester, such as di-(2-ethylhexyl)phthalate, dioctyl phthalate, dibutyl phtalate, phthalic acid benzyl butyl ester, and phthalic acid diisononyl ester Aliphatic-carboxylic-acid ester, such as dioctyl adipate, a succinic acid screw (2-methylnonyl), dibutyl sebacate, and butyl oleate. Alcohol ester, such as pentaerythritol ester

[0055]Phosphoric ester, such as trioctyl phosphate and tricresyl phosphate Epoxy plasticizers, such as epoxidized soybean oil, 4,5-epoxy hexahydrophthalic acid dioctyl, and epoxy stearic acid benzyl. Chlorinated paraffin. Polyester plasticizers, such as polyester dibasic acid and dihydric alcohol are made to come to reset.

[0056]Polyoxyalkylene, polyester, Poliy alpha-methylstyrene, Polymeric plasticizers, such as polystyrene, polybutadiene, alkyd resin, polychloroprene, polyisoprene, polybutene, hydrogenation polybutene, epoxidation polybutadiene, and Butadiene Acrylonitrile. The viewpoint of a polyoxyalkylene polymer (A) and compatibility with (B) to polyoxyalkylene is [among these] preferred. As such polyoxyalkylene, 1.5 or less polyoxyalkylene has preferred Mw/Mn which a with a molecular weights of 4000 or more thing was preferred, and was manufactured using the compound cyanide complex etc. from the point that a low molecular weight body is hypoviscosity few. As such polyoxyalkylene, hydroxyl group content polyoxyalkylene may be sufficient and the polymer produced by changing the hydroxyl group into other organic groups may be sufficient. Especially the thing changed into the polymer which specifically closed terminal hydroxyl groups by hydrocarbon groups, such as an alkyl group and an alkenyl group, via combination of an ether bond, an ester bond, a urethane bond, etc. is preferred. It is the polymer most preferably closed by the allyl group via the ether bond.

[0057]These plasticizers can be suitably chosen according to a use or the purpose. Although use of high di-(2-ethylhexyl)phthalate of flexibility is the most common, For example, when high weatherability wants to improve, an adhesive property can be improved by considering it as the non-plastic combination with a large molecular weight uses what is called a polymeric plasticizer, and a plasticizer does not use a plasticizer rather rather than is necessarily required in the case of an adhesives use.

[0058]These plasticizers may be used independently and may use two or more sorts together. (Solvent) When using the constituent of this invention as a hardenability constituent again, when adjusting viscosity, in the non-plastic combination which can also add a solvent for the purpose of adjustment of viscosity, and the improvement in preservation stability of a constituent, and does not mainly use plasticizers, such as an adhesives use, in particular, it is useful. The amount of the solvent used has 0.001 - 500 preferred weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B).

[0059]As a solvent, aliphatic hydrocarbon, aromatic hydrocarbon, and halogenated hydrocarbon. Alcohols, ketone, ester species, ether, ester alcohols, ketone alcohol, ether alcohol, ketone ether, ketone ester species, and ester ether can be used. When saving the constituent of this invention at a long period of time, since preservation stability of alcohols improves, they are preferred. As alcohols, alkyl alcohol of the carbon numbers 1-10 is preferred, and methanol, ethanol, isopropanol, isopentyl alcohol, especially hexyl alcohol, etc. are preferred.

(Hardening accelerator catalyst) When stiffening the hardenability constituent in this invention, the hardening accelerator catalyst which promotes the hardening reaction of a hydrolytic basis content silicon group may be used. The following compound is mentioned as a concrete example. Those one sort or two sorts or more are used. As for a hardening accelerator catalyst, it is preferred to carry out 0.0001-10 weight-section use to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B).

[0060]Specifically, the tin compound shown below is mentioned. Various compounds of divalent tin, such as 2-ethylhexanoic acid tin, naphthenic acid tin, and stearic acid tin. Dibutyltin dilaurate, dibutyltin diacetate, dibutyltin mono- acetate, Organic tin carboxylate like dialkyl tin dicarboxylates, such as dibutyltin malate, or dialkyl tin mono- carboxylate, The Suzuki rate compounds, such as dialkyl tin bisacetylacetonate and a JIARUKIRUSUZU monoacetyl acetonato monoalkoxide, Tetravalent tin various compounds, such as a reactant of dialkyl tinoxide, the reactant of an ester compound and dialkyl tinoxide, and an alkoxy silane compound, and a dialkyl tin dialkyl sulfide.

[0061]As a Suzuki rate compound, dibutyltin bisacetylacetonate, dibutyltin bis-ethylacetoacetate, a dibutyltin monoacetyl acetonate monoalkoxide, etc. are mentioned. Carry out heating mixing,

dibutyltin oxide and phthalic ester, such as dioctyl phthalate and phthalic acid diisononyl ester, are made to react as a reactant of dialkyl tinoxide and an ester compound, and the tin compound made liquefied is mentioned. In this case, as an ester compound, tetraethyl silicate, its partial hydrolysis condensate, etc. can be used besides aster of aliphatic series and aromatic carboxylic acid.

[0062]The compound which reacted or mixed these tin compounds with low molecule alkoxysilane etc. is also preferred, and can be used. The following are mentioned as a hardening accelerator which can be used besides a tin compound. Metal salt, such as an alkyl titanate, an organic silicon titanate, organic-carboxylic-acid bismuth salt, and organic-carboxylic-acid lead salt.

[0063]Acidic compounds, such as phosphoric acid, p-toluenesulfonic acid, phthalic acid, and a phosphoric acid ester (2-ethylhexyl). A butylamine, hexylamine, octylamine, decyl amine, lauryl amine, Aliphatic series monoamines, such as N,N-dimethyl- octylamine, ethylenediamine, Hexamethylenediamine, diethylenetriamine, triethylenetetramine, Amine compounds, such as aminosilane coupling agents, such as aliphatic polyamine compounds, such as tetraethylenepentamine, an aromatic amine compound, alkanolamine, 3-(2-aminoethyl) amino-propyltrimethoxysilane, and 3-aminopropyl trimethoxysilane.

[0064]If a tin compound and a bismuth compound are used together with an amine compound, especially the 1st class amine compound, since their hardening facilitatory effect will improve, their concomitant use is preferred. By combining basic compounds, such as an above-mentioned acidic compound, an amine compound, etc., shows a higher hardening facilitatory effect in a high temperature region comparatively especially. A hardening accelerator can be used combining one sort or two sorts or more.

(Storage stabilizer) A storage stabilizer can be added in order to improve the storage stability of the hardenability constituent of this invention further again. A storage stabilizer means the compound which can control hydrolysis of a polyoxyalkylene polymer (A) and (B) under existence of a small amount of water, or a compound with the high drying effect.

[0065]The amount of the storage stabilizer used has 0 - 30 preferred weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B). A compound which generates strong acid by hydrolysis like mineral acid or chlorosilicane demonstrates the effect by one or less weight section. Specifically, the following compounds are raised. It is preferred to be simultaneously added at least, before using independently also being able to use these together and adding a curing catalyst.

[0066]Methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, isobutyl alcohol, 2-butanol, t-butyl alcohol, n-amyl alcohol, Alcohols, such as isoamyl alcohol, 1-hexanol, octyl alcohol, 2-ethylhexanol, cellosolve, ethylene glycol, propylene glycol, glycerin, a diethylene glycol, and dipropylene glycol.

[0067]Malonic ester compounds, such as 1,3-diketones, such as an acetylacetone, methyl acetoacetate, ethyl acetoacetate, and butyl acetoacetate, and an acetoacetic ester compound, dimethyl malonate, and diethyl malonate etc. An mineral acid, it is minaral acid, such as ohloride, sulfuric acid, and nitric acid, etc. $(Me_2CHO)_2P(=O)(OH)$ $(Me_2CHO)_2P(=O)(OH)_2$, $(EtO)_2P(=O)(OH)$ $(EtO)P(=O)(OH)_2$, (MeO) Phosphoric acid alkyl ester, such as $_2P(=O)(OH)$ $(MaO)P(=O)(OH)_2$ and $(BuO)_2P(=O)(OH)$ $(BuO)P(=O)(OH)_2$ and $(PrO)_2P(=O)(OH)$ $(PrO)P(=O)(OH)_2$(In addition, a methyl group and Et express an ethyl group, Pr expresses a propyl group, Bu expresses a butyl group, and Me is the same as that of below.)

ORUTOGI acid alkyl ester, such as ORUTOGI acid trimethyl ** triethyl orthoformate, tripropyl orthoformate, and ORUTOGI acid tributyl

[0068]As alt.acetic acid alkyl ester, it is alt.acetic acid alkyl ester, such as alt.acetic acid trimethyl ** alt.acetic acid triethyl. Tetrasilkyl silicate, such as tetramethyl silicate, tetraethyl silicate, tetrapropylsilicate, and tetrabutyl silicate. The partial hydrolysis condensate of tetramethyl silicate and/or tetraethyl silicate.

[0069]Alkyltrialkoxysilane, such as methyl trimetoxysilane and ethyltrimethoxysilane. Alkenyl trialkoxysilane, such as vinyltrimetoxysilane. Aryl trialkoxysilane, such as phenyltrimethoxysilane. Chlorosilicanes, such as methyltrichlorosilane, methyldi chlorosilicane, dimathyldichlorosilane, trimethylchlorosilane, phenyltrichlorosilana, and diphenyl dichlorosilane

[0070]Silazanes, such as hexamethyldisilazane

$CH_2=Si(ON=CMe_2)_2$, $MeSi(NMe_2)_3$, $MeSi(ONMe_2)_3$, $MeSi(NMaC(=O)Me)_3$, $MeSi(OCMe=CH_2)_3$,

[0071]Hydrolytic organic titanium compounds, such as tetraisopropoxy titanium, tatrabutoxytitanium, and tetraoctyl oxytitanium, And those condensates, $Ti(OCHMe_2)_2[OGMe=OHCOMe]_2$, $Ti(OBu)_2[OC_2H_4N(C_2H_4OH)_2]_2$, $Ti(OH)_2[OOHMeCOOH]_2$, $Ti(OCHMe_2)_3[OOC_{17}H_{35}]$, $Ti(OCHMe_2)[OOOC_{17}H_{35}]_3$, isopropoxy titanium dimethacrylate monoisostearate, isopropoxy titanium tris (4-amino benzoate), isopropoxy titanium tris (dioctyl phosphate), etc.

[0072]Zeolite. Alkyl mercaptan, such as dodecyl mercaptan and t-butyl mercaptan. Mercaptosilanes, such as gamma-mercapto propyltrimethoxysilane. Sulfhydryl group content carboxylic acid, such as 2-mercaptopropionic acid and thiosalicylic acid.

[0073]Sulfhydryl group content ester compounds, such as 2-ethylhexyl thioglycolate. Sulfhydryl group content polymer like KAPUKYUA 3-800 (Diamond SHAMUROKKU Chemicals both-ends sulfhydryl group content polyoxyalkylene), a thiophenol, thiobenzoic acid, etc.

[0074]The partial hydrolysis condensate of the point of cost and an effect to vinyltrimetoxysilane, a tetraethoxysilane, tetramethyl silicate, and/or tetraethyl silicate is [among these] the most preferred.

(Thixotropy grant agent) A thixotropy grant agent may be used again for the improvement of lappet nature. As such a thixotropy grant agent, hydrogenation castor oil, fatty acid amide, etc. are used.

(Antiaging agent) As an antiaging agent again A hindered amine system, a benzotriazol system, Antioxidants generally used, such as each compound of a benzophenone series, a benzoate system, a cyanoacrylate system, an acrylate system, a hindered phenol system, the Lynn system, and sulfur systems, light stabilizer, and an ultraviolet ray absorbent are used suitably. These antiaging agents have especially a preferred thing that it may use together and is used together. It is desirable method from being in the case of raising an effect as a whole taking advantage of each feature by using it especially combining two sorts or all from light stabilizer, an antioxidant, and an ultraviolet ray absorbent. Specifically, especially the thing for which the hindered amine light stabiliser of the 3rd class or the 2nd class, a benzotriazol system ultraviolet ray absorbent, a hindered phenol system, and/or a phosphite system antioxidant are combined is effective. As for the amount of the antiaging agent used, it is preferred that it is the range of 0.1 - 10 weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B), respectively. In less than 0.1 weight sections, there are few effects of an improvement of weatherability, and if five weight sections are exceeded, there is no great difference in an effect and it is economically disadvantageous.

[0075]As an antioxidant, a hindered phenol system and/or a phosphite system anti-oxidant are preferred, and, specifically, the following can be illustrated. NOKURAKKU 200, NOKURAKKU M-17, NOKURAKKU SP, NOKURAKKU SP-N, NOKURAKKU NS-5, NOKURAKKU NS-6, NOKURAKKU NS-30, NOKURAKKU 300, NOKURAKKU NS-7, NOKURAKKU DAH (all are the products made from the Ouchi Shinko Chemical industry above), ADEKA stub AO-30, ADEKA stub AO-40, ADEKA stub AO-50, ADEKA stub AO-60, ADEKA stub AO-616, ADEKA stub AO-635, ADEKA stub AO-18, the ADEKA stub 328, ADEKA stub AO-37. (-- the above -- any --) by Asahi Denka Kogyo K.K.. IRGANOX-245, IRGANOX-259, IRGANOX-565, IRGANOX-1010, IRGANOX-1035, IRGANOX-1076, IRGANOX-1081, IRGANOX-1098, IRGANOX-1222, IRGANOX-1330, IRGANOX-1425WL (all are made in Tiba Specialty Chemicals, Inc. above).

[0076]As light stabilizer, the 2nd class and/or the 3rd class hindered amine system compound have them, and specifically, [preferred] Tinuvin 622LD, the tinuvin 144, CHIMASSORB944LD, CHIMASSORB119floor line (all are made in Tiba Specialty Chemicals, Inc. above), ADEKA stub LA-57, ADEKA stub LA-62, ADEKA stub LA-67, ADEKA stub LA-63P, ADEKA stub LA-68LD, ADEKA stub LA-601 (all are the Asahi Denka Kogyo K.K. make above), SANORU LS-770, SANORU LS-765, SANORU LS-292, SANORU LS-2626, SANORU LS-1114, SANORU LS-744 (all are the Sankyo Co., Ltd. make above), etc. can be illustrated.

[0077]As an ultraviolet ray absorbent, a benzotriazol system compound or a benzoate system compound has it, and specifically, [preferred] The tinuvin P, the tinuvin 234, the tinuvin 320, the tinuvin 326, the tinuvin 327, the tinuvin 329, the tinuvin 213, and the tinuvin 120 (all are made in Tiba Specialty Chemicals, Inc. above) can be illustrated.

[0078]The tinuvin B5353 which is a mixture of the above-mentioned compound, the tinuvin B75 (all are made in Tiba Specialty Chemicals, Inc. above), etc. can be used.

(A curing-in-air compound, a photoresist compound) An air-oxidation hardenability compound and a

photoresist compound can be added in order to improve the adhesion of dust, and surface tackiness over a long period of time. Using together is more preferred although these compounds may be used independently. The amount used has 0.001 - 50 preferred weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B).

[0079]The compound which contains in intramolecular tha unsaturation group which causes a reaction by oxygen in the air as an air-oxidation hardensability compound is preferred. Specifically Drying oil, such as tung oil, linseed oil, perilla oil, soybean oil, sunflower seed oil, and hempseed oil. The resultant of the various alkyd resins and drying oil which are produced by carrying out conversion of the drying oil and functionality polyoxyalkylene. The resultant (urethane oil) of drying oil and an isocyanate compound, the acrylic polymer by which conversion was carried out with drying oil. The epoxy resin by which conversion was carried out with drying oil, the silicon resin by which conversion was carried out with drying oil, Diene system polymers, such as a polymer of polybutadiene and diene of the carbon numbers 5-8, and a copolymer, the allyloxy group content polyester compound (air-drying nature unsaturated polyester) obtained by the polycondensation of allyloxy group content glycol and polyvalent carboxylic acid -- the various denaturation things (mallein-ized denaturation, boiled oil denaturation, etc.) of this polymer or a copolymer, etc. are mentioned further.

[0080]As a photoresist compound, what molecular structure causes a chemical change considerably for a short time, and produces physical-properties change of hardening etc. by operation of light can be used. Many things, such as a constituent which contains a monomer, oligomer, resin, or them as this kind of a compound, are known and marketed, and can use these publicly known compounds arbitrarily. Among these Polyhydric alcohol, such as trimethylolpropane, polyether polyol, Polyfunctional acrylate, such as a compound containing the acrylyl group produced by making a hydroxy compound, acrylic acid, and methacrylic acid, such as polyester polyol, react (meta), is used the general-purpose.

(Modulus regulator) The compound which has one silanol group in intramolecular, or the compound which can generate the compound which has one silanol group in intramolecular can be added for physical-properties adjustment of a hardened material and the purpose of surface stickiness reduction again. The effect of reducing a modulus by addition of these compounds without worsening stickiness of the surface is acquired. The amount used has 0 - 10 preferred weight section to a polyoxyalkylene polymer (A) and a total of 100 weight sections of (B).

[0081]As a compound which has one silanol group, a trimethyl silanol, triethyl silanol, a triphenylsilanol, etc. are mentioned to intramolecular. As a compound which can generate the compound which has one silanol group, trimethylsilyl ether, such as fatty alcohol and aromatic alcohol, can be used for intramolecular. Specifically N-trimathylsilyl acetamide, hexamethyldisilazane, Methoxy trimethylsilane and athoxy trimethylsilane, isopropyloxy trimethylsilane, Butoxy trimethylsilane, hexyloxy trimethylsilane, 2-ethylhexyloxy silane, n-octyloxy silane, 2-chloropropyl oxy trimethylsilane, Phenoxy trimethylsilane, 2-methylphenoxy trimethylsilane, 2-chlorophenoxy trimethylsilane, methoxy methyl phenylsilane, methoxy methylal phenylsilane, phenoxydimethyl phenylsilane, etc. are mentioned. Ethylene glycol, propylene glycol, 1,3-propanediol, Dipropylene glycol, 1,2-butanediol, 1,4-butanediol, The compound etc. which carried out trimethylsilyl etherification of the hydroxyl group of multivalent hydroxy compounds, such as 1,6-hexanediol, glycerin, trimethylolpropane, pentaerythritol, and sorbitol, can be used.

(In addition to this) Organic colors, such as inorganic pigments, such as iron oxide, chrome oxide, and titanium oxide, and copper phthalocyanine blue, and Phthalocyanine Green, can be used as paints.

[0082]According to a use, publicly known additive agents, such as mildewproofing material and fire retardant, can be used. The room-temperature-curing nature constituent of this invention is suitable for the use as which it can be used for sealant, a water blocking material, adhesives, a coating agent, etc., and the dynamic flattery nature to the sufficient cohesive force and adherend of especially the hardened material itself is required.

[0083]

[Example]The example and comparative example of this invention are explained below. A part shows a weight section. In the example 1-1 to 1-6 of manufacture, a hydroxyl value conversion molecular weight shows the molecular weight converted from the hydroxyl value of the polyoxyalkylene polymer which has a hydroxyl group which is a raw material. Mw/Mn is the value measured by the gel

permeation chromatograph using a tetrahydrofuran as a solvent. The analytical curve was prepared using the correlation sample of styrene.

(Example 1-1 of menufacture) Use glycerin as an initiator and Under existence of a zinchexacyano cobaltate glyme complex compound catalyst, To the hydroxyl value conversion molecular weight 17000 produced by making propylene oxide react, and polyoxypropylene triol of Mw/Mn=1.3. The methanol solution of sodium methoxide was added, scale loss pressing-down methanol was distilled off, and the terminal hydroxyl groups of polypropylene oxide were changed into sodium alcoholate. Next, the allyl chloride was made to react, the unreacted allyl chloride was removed and refined, and the polypropylene oxide which has an allyl group at the end was obtained. it was alike, received this reactant, methyl dimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst, and the polypropylene oxide (P1) which has a methyl dimethoxy silyl group at the end was obtained.

(Example 1-2 of manufacture) Use propylene glycol as an initiator and Under existence of a zinchexacyano cobaltate glyme complex compound catalyst, The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained using the hydroxyl value conversion molecular weight 17000 produced by making propylene oxide react, and polyoxypropylene diol of Mw/Mn=1.3. Methyl dimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to this reactant, and the polypropylene oxide (P2) which has a methyl dimethoxy silyl group at the end was obtained.

(Example 1-3 of menufacture) The methanol solution of sodium methoxide was added to polyoxypropylene diol of the hydroxyl value conversion molecular weight 3000 obtained using the potassium hydroxide catalyst, scale loss pressing-down methanol was distilled off to it, and terminal hydroxyl groups was changed into it at sodium alcoholate. Next, after making it react to chlorobromomethane and performing polymers quantification, the allyl chloride was made to react continuously, it refined, and the polypropylene oxide (Mw/Mn=2.0) which has an allyloxy group at the end was obtained. Methyl dimethoxysilane which is a hydrosilyl compound was made to react to this under existence of a platinum catalyst, and the polypropylene oxide (P3) of the molecular weight 9000 which has a methyl dimethoxy silyl group at the end was obtained.

(Example 1-4 of manufacture) Use glycerin as an initiator and Under existence of a zinchexacyano cobaltate glyme complex compound catalyst, The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained using the hydroxyl value conversion molecular weight 17000 produced by making propylene oxide react, and polyoxypropylene triol of Mw/Mn=1.3. To this reactant, 3-mercapto propyltrimethoxysilane which is a silyl compound was made to react using the 2,2'-azobis 2-methylbutyronitrile which is a polymerization initiator, and the polypropylene oxide (P4) which has a trimethoxysilyl group at the end was obtained.

(Example 1-5 of manufacture) Use propylene glycol as an initiator and Under existence of a zinchexacyano cobaltate glyme complex compound catalyst, The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained using the hydroxyl value conversion molecular weight 17000 produced by making propylene oxide react, and polyoxypropylene diol of Mw/Mn=1.3. To this reactant, 3-mercapto propyltrimethoxysilane which is a silyl compound was made to react using the 2,2'-azobis 2-methylbutyronitrile which is a polymerization initiator, and the polypropylene oxide (P5) which has a trimethoxysilyl group at the end was obtained.

(Example 1-6 of manufacture) It refined, after having used glycerin as the initiator, performing the polymerization of propylene oxide under existence of a zinchexacyano cobaltate glyme complex compound catalyst and obtaining the hydroxyl value conversion molecular weight 10000 and polyoxypropylene triol of Mw/Mn=1.2. gamma-isocyanate propyltrimethoxysilane was added to this, the urethane-ized reaction was performed, and the polypropylene oxide (P6) which has a trimethoxysilyl group at the end was obtained.

(Example 2-1 of manufatoure) Use polymeric plasticizer glycerin as an initiator and Under existence of a zinchexacyano cobaltate glyme complex compound catalyst, The polypropylene oxide (G1) which has an allyl group at the end by the same method as the example 1 of manufacture was obtained using the hydroxyl value conversion molecular weight 17000 produced by making propylene oxide react, and polyoxypropylene triol of Mw/Mn=1.3.

(Example 2-2 of manufacture) It refined, after having used polymeric plasticizer glycerin as the

initiator, performing the polymerization of propylene oxide under existance of a zinchexacyano cobaltate glyme complex compound catalyst and obtaining the hydroxyl value converaion molecular weight 10000 and polyoxypropylene triol of Mw/Mn=1.2. (G2)

(Example 3-1 of manufacture) Add 0.2 mol of dibutyltin oxide to toluene 150cm^3 in polymer hardening accelerator catalyst glass reactors, and 0.1 mol of 2-ethylhexanol is added, It was made to react removing the water which oarries out azeotropy to bottom toluene of heating chuming until the water of the amount of theories finishes distilling off. Then, 0.1 mol of acetylacetones are added, and it was made to react removing the water which carries out azeotropy to toluene further until the water of the amount of theories finishes distilling off. It filtered in order to remove a little sediments, and it distilled off under decompression of toluene further, and the light yellow tin compound (J1) was obtained.

(Example 3-2 of manufacture) In the 3 Thu mouth flask of the glass furnished with a polymer hardening accelerator catalyst flowing-back condenser tube and an agitator. 1 mol of dibutyltin oxide and 0.5 mol of 2-ethylhexanoic acid ethyl were added, and it heated at 120 ** for 5 hours, agitating under a nitrogen atmosphere, and the tin compound (J2) of uniform light yellow was obtained.

(Example 4-1 of manufacture) Weighing of epicure H-1(epoxy resin hardener by oil recovery shell epoxy company)2 mol and the KBM403 (epoxysilane by Shin-etsu chemicals company) 2.5 mol is carried out to epoxy curing agent glass reactors, The pyrogenetic reaction was carriad out at 90 ** for 5 hours, agitating under a nitrogen atmosphere, and the compound (K1) was obtained.

(Example 5-1 of manufactura) 1 mol of trimethylolpropane was put into the glass reaction vessels which attached the modulus regulator dropping funnel, the flowing-baok condenser tube, and the agitator, and 3.3 mol of sodium hydroxide ground further was added. 3.0 mol of trimethylohlorosilanes were dropped over 1 hour from the dropping funnel at the bottom room temperature of churning. Temperature up was carried out to 40 ** after the end of dropping, and it agitated for 8 hours. Fine sodium chloride has deposited in a reaction vessel as a reaction progresses. By analyzing a trimethylsilyl group with a nuclear magnetic resonance spectrum, After the saturation sodium chloride aqueous solution washed after washing enough after checking that the reaction had been completed mostly until it filtered the reaction mixture, the water layer checked filtrate with the pH test paper in order of ammonium chloride solution and ion exchange water and it became pH7 mostly, anhydrous sodium sulfate was put into the organic layer, and it dried to it. Filtration removed anhydrous sodium sulfate, distillation refining was carried out under decompression, and the trimethylsilyl ether (L1) of trimethylolpropane was obtained.

(Example 5-2 of manufacture) To the glass reaction vessels which attached the modulus regulator dropping funnel, the flowing-back condenser tube, and the agitator. 1.5 mol of 2-ethylhexyl alcohol was put in, and 0.5 mol of trimethylohlorosilane and the mixture of 0.5 mol of hexamethyldisilazane were dropped over 2 hours under churning at the room temperature. It agitated at 40 ** after the end of dropping for 5 houra, 0.2 more mol of methanol was added, and it allowed to stand at the room temperature all night. After it removed tha ammonium ohloride which carried out the byproduction by filtration on the next day, and ammonium chloride solution washed filtrate and drying and filtering with anhydrous sodium sulfate, distillation refining was carried out under decompression and the trimethylsilyl ether (L2) of 2-ethylhexyl alcohol was obtained.

(Example 1 of an examination) Polypropylene oxide (P1) - (P6) manufactured in the example 1-1 to 1-6 of manufacture and an antiaging agent, and a thixotropy grant agent to what carried out mixture dispersion. The bulking agent and paints from which moisture was beforehand removed by stoving were added, and it mixed, and Epicoat 828 (bisphenol A type epoxy resin by an oil recovery shell epoxy company), the epoxy curing agent, the plasticizer, and the storage stabilizer were added further, and it mixed. Furthermore the silane compound and the curing catalyst were added, mixing and after defoaming, the cartridge for sealing materials which can interrupt penetration of the hygroscopic surface moisture in the air was filled up, and the constituents D1-D8 of this invention and the comparative constituents D9-D12 were obtained. The kind and quantity of each raw material are as having been shown in Table 1.

[0084]three-stage evaluation of the ease (handling nature) of dealing with it of a class product was carried out (O which is especially easy to deal with it, :O which ls easy to deal with it, :x which is hard to deal with it, and * — the thing which has the low viscosity of a constituent is dealt with generally, and a sex is good). A result is shown in Table 1-1.

[0085]
[Table 1]

表1-1

			実施例								比較例			
		組成物番号	D1	D2	D3	D4	D5	D6	D7	D8	D9	D10	D11	D12
配合	ポリプロピレンオキシド	P1	56.3	37.5	18.8	37.5				25		75		50
		P2					37.5	37.5						
		P3							37.5		37.5			
		P4	18.8	37.5	56.3		37.5		37.5	25			75	50
		P5				37.5		37.5						
		P6									37.5			
	エポキシ樹脂 (C)	エピコート828 *1	25	25	25	25	35	25	25	50	25	25	25	
	エポキシ硬化剤 (E)	H-3 *2	4	4	4	4				8	4	4	4	
		X1					9	9	9					
	充填剤	ネオライトSP-T *3	50	50	50	50	50	50	50	50	50	50	50	50
		ホワイトンSS *4	100	100	100	100	100	100	100	100	100	100	100	100
		中空体1 *5				1								
		中空体2 *6					1							
		中空体3 *7						1						
		中空体4 *8							1					
	顔料	R820 *9	5	5	5	5	5	5		5	5	5	5	5
	可塑剤	ハイゾール *10	10	10	10	10				10	10	10	10	10
		G1					25							
		G2						25						
	チキソ性付与剤	ディスパロン6500 *11	3	3	3					3	3	3	3	3
	シラン化合物 (F)	アミノシラン1 *12				1			0.5					
		アミノシラン2 *13					1							
		アミノシラン3 *14						1						
		エポキシシラン *15							0.5	1	1	1	1	1
	貯蔵安定剤	VTMS *16	2	2	2					2	2	2	2	2
		TES *17				2	3	2	2					
	老化防止剤	アデカスタブLA62 *18					1		0.5					
		アデカスタブLA63P *19						1	0.5					
		チヌビン327 *20	1	1	1	1	1	1	1	1	1	1	1	1
		イルガノックス1010 *21	1	1	1	1	1	1	1	1	1	1	1	1
	硬化触媒	AC *22				1								
		J1					1							
		J2						1						
		J3 *23	1	1	1				1	1	1	2	1	1
		DBTDL *24							0.1					
試験結果		取り扱い性	○	○	○	○	○	○	○	○	×	○	○	○

*1 ビスフェノールA型エポキシ樹脂（油化シェルエポキシ）
*2 エピキュアH-1（油化シェルエポキシ）
*3 軽質炭酸カルシウム［竹原化学工業（株）］
*4 重質炭酸カルシウム［白石カルシウム工業（株）］
*5 CEL-STAR SX-39（東海工業）
*6 ユーカーフェノリックマイクロバルーンズ BJO-0930（ユニオンカーバイド）
*7 マツモトマイクロスフェアーMFL-100CA（松本油脂製薬）
*8 マツモトマイクロスフェアーMFL-80GCA（松本油脂製薬）
*9 酸化チタン（石原産業）
*10 酸化水素添加芳香族炭化水素（日本石油化学）
*11 脂肪酸アミド（楠本化成）
*12 3-アミノプロピルトリメトキシシラン
*13 N-（2-アミノエチル）-3-アミノプロピルトリメトキシシラン
*14 N-（2-アミノエチル）-3-アミノプロピルメチルジメトキシシラン
*15 3-グリシドキシプロピルトリメトキシシラン
*16 ビニルトリメトキシシラン
*17 テトラエトキシシラン
*18 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*19 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*20 ベンゾトリアゾール系光安定剤（チバ・スペシャルティ・ケミカルズ株式会社）
*21 ヒンダードフェノール系酸化防止剤（チバ・スペシャルティ・ケミカルズ株式会社）
*22 ジブチルスズビスアセチルアセトナート
*23 ジブチルスズオキシドと酢酸との反応物
*24 ジブチルスズジラウレート

[0086]The following examinations were done about the constituent (11 points) which was good among the obtained constituents as for handling nature. A result is shown in Table 1-2.

(Tuck free examination) The tuck free examination was done based on JIS A5758. The class product was extruded from the cartridge, and promptly, on the glass plate, the spatula was used for about 3 mm in thickness, and it accustomed level, and placed at the normal condition. The time which it took that a constituent stops adhering to a fingertip since the surface was lightly touched and accustomed it by the fingertip purified by ethanol level was measured, and it was considered as tack free time.

(Tension shear test) In [extrude a class product from a cartridge and] the bottom of 50% of humidity at 23 **, To an aluminum plate (25 mm in width, 100 mm in length, and 3 mm in thickness), an adhesion area of 25 mm x 25 mm, It applied so that it might be set to about 0.2 mm in thickness, and in 30 seoonds, one more piece of aluminum plate is piled up alternately, and was stuck, and tensile shear bond strength (unit: kg/cm^2) was meaured after care of health for seven days on the conditions with 5-mm the speed of teating for /.

[0087]

[Table 2]

表1－1

区分	成分	組成物番号	D1	D2	D3	D4	D5	D6	D7	D8	D10	D11	D13
			実施例								比較例		
配合	ポリプロピレンオキシド	P1	56.3	37.5	18.8	37.5				25	75		37.5
		P2					37.5	37.5					
		P3							37.5				
		P4	18.8	37.5	56.3		37.5		37.5	25		75	37.5
		P5				37.5		37.5					
		P6											
	エポキシ樹脂（C）	エピコート828 *1	25	25	25	25	25	25	25	50	25	25	
	エポキシ硬化剤（E）	H-3 *2	4	4	4	4				6	4	4	
		K1					9	9	9				
	充填剤	ネオライトSP-T *3	50	50	50	50	50	50	50	50	50	50	50
		ホワイトンSB *4	100	100	100	100	100	100	100	100	100	100	100
		中空体1 *5				1							
		中空体2 *6					1						
		中空体3 *7						1					
		中空体4 *8							1				
	顔料	R820 *9	5	5	5	5	5	5		5	5	5	5
	可塑剤	ハイゾール *10	10	10	10	10				10	10	10	10
		G1					25						
		G2						25					
	チキソ性付与剤	ディスパロン6500 *11	3	3	3					5	3	3	3
	シラン化合物（F）	アミノシラン1 *12				1			0.5				
		アミノシラン2 *13					1						
		アミノシラン3 *14						1					
		エポキシシラン *15							0.5	1	1	1	2
	貯蔵安定剤	VTMS *16	2	1	1					2	3	2	2
		TES *17				2	2	2	2				
	老化防止剤	アデカスタブLA62 *18					1		0.5				
		アデカスタブLA63P *19						1	0.5				
		チヌビン327 *20	1	1	1	1	1	1	1	1	1	1	1
		イルガノックス1010 *21	1	1	1	1	1	1	1	1	1	1	1
	硬化触媒	AC *22				1							
		J1					1						
		J2						1					
		J3 *23	1	1	1				1	1	1	1	1
		DBTDL *24							0.2				
試験結果		タックフリータイム[hr]	2.2	1.4	0.8	1.0	1.8	1.2	3.4	3.1	40	0.6	1.2
		引張剪断接着強さ[kg/cm²]	37	33	34	36	38	37	30	30	35	32	7

*1 ビスフェノールA型エポキシ樹脂（油化シェルエポキシ）
*2 エピキュアH－3（油化シェルエポキシ）
*3 膠質炭酸カルシウム（竹原化学工業（株））
*4 重質炭酸カルシウム（白石カルシウム工業（株））
*5 CEL-STAR SX-39（東海工業）
*6 ユーカーフェノリックマイクロバルーンズ BJO-0930（ユニオンカーバイド）
*7 マツモトマイクロスフェアーMFL-100CA（松本油脂製薬）
*8 マツモトマイクロスフェアーMFL-80GCA（松本油脂製薬）
*9 酸化チタン（石原産業）
*10 炭化水素系高沸点溶剤（日本石油化学）
*11 脂肪酸アミド（楠本化成）
*12 3－アミノプロピルトリメトキシシラン
*13 N－（2－アミノエチル）－3－アミノプロピルトリメトキシシラン
*14 N－（2－アミノエチル）－3－アミノプロピルメチルジメトキシシラン
*15 3－グリシドキシプロピルトリメトキシシラン
*16 ビニルトリメトキシシラン
*17 テトラエトキシシラン
*18 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*19 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*20 ベンゾトリアゾール系光安定剤（チバ・スペシャルティ・ケミカルズ株式会社）
*21 ヒンダードフェノール系酸化防止剤（チバ・スペシャルティ・ケミカルズ株式会社）
*22 ジブチルスズビスアセチルアセトナート
*23 ジブチルスズオキシドと酢酸との反応物
*24 ジブチルスズジラウレート

[0088](Example 2 of an examination) A plasticizer, a bulking agent, and paints were added to Epicoat 828 (bisphenol A type epoxy resin by an oil recovery shell epoxy company), it mixed to it, and this was made into A agent. To what added the polypropylene oxide (P1) manufactured in the example 1-1 to 1-6 of manufacture - (P6) the modulus regulator, and the curing-in-air compound, was mixed, and added and carried out mixture dispersion of an antiaging agent and the thixotropy grant agent further. The bulking agent and paints from which moisture was beforehand removed by stoving were added, and it mixed, and the plasticizer, the storage stabilizer, and the epoxy curing agent were added further, and it mixed. Furthermore the silane compound and the curing catalyst were added, mixing and defoaming of were done, and this was made into B agent. The kind and quantity of each raw material are as having been shown in Table 2.

[0089]A agent and B agent which were obtained were mixed and agitated just before use, and the constituents D13-D20 of this invention and the comparative constituent D21 - 24 were obtained. three-stage evaluation of the ease (handling nature) of dealing with it of a class product was carried out (O which is especially easy to deal with it, :O which is easy to deal with it, :x which is hard to deal with it, and * — the thing which has the low viscosity of a constituent is dealt with generally, and a sex is good). A result is shown in Table 2-1.

[0090]
[Table 3]

表2－1

区分	成分	組成物番号	D13	D14	D15	D16	D17	D18	D19	D20	D21	D22	D23	D24
			実施例								比較例			
配合（A剤）	エポキシ樹脂（C）	エピコート828 *1	25	25	25	25	25	25	25	50	25	25	25	
	充填剤	ネオライトSP-T *3	20	20	20	20	20	20	10	20	20	20	20	
		ホワイトンSB *4	45	45	45	45	45	45	45	45	45	45	45	
	顔料	R820 *9	5	5	5	5	5	5		5	5	5	5	
	可塑剤	ハイゾール *10	5	5	5	5	5	5	5		5	5	5	
（B剤）	ポリプロピレンオキシド	P1	56.3	37.5	18.8	37.5				25		75		50
		P2					37.5	37.5						
		P3							37.5		37.5			
		P4	18.8	37.5	56.3		37.5		37.5	25			75	50
		P5				37.5		37.5						
		P6									37.5			
	充填剤	ネオライトSP-T *3	30	30	30	30	30	30	30	30	30	30	30	50
		ホワイトンSB *4	60	60	60	60	60	60	50	60	60	60	60	100
		中空体1 *5				1								
		中空体2 *6					1							
		中空体3 *7						1						
		中空体4 *8							1					
	可塑剤	ハイゾール *10	5	5	5					5	5	5	5	10
		G1					20							
		G2						20						
		DOP *25				5								
	チキソ性付与剤	ディスパロン6500 *26	3	3	3					3	3	3	3	3
	シラン化合物（F）	アミノシラン1 *12	1	1	1	1	1	1	1	1	1	1	1	1
	貯蔵安定剤	VTMS *16	2	1	2					2	2	2	2	2
		TES *17				2	2	2	2					
	老化防止剤	アデカスタブLA62 *18					1		0.5					
		アデカスタブLA63P *19						1	0.5					
		チヌビン327 *20	1	1	1	1	1	1	1	1	1	1	1	1
		イルガノックス1010 *21	1	1	1	1	1	1	1	1	1	1	1	1
	モジュラス調整剤	L1					0.7		0.4					
		L2				1.3								
		L3 *27							0.4					
		L4 *28						0.5						
	空気硬化性化合物	桐油	5	5	5	5	5	5		5	5	5	5	5
	光硬化性化合物	アロニックスM309 *29	3	3	3		3	3	10	3	3	3	3	3
	エポキシ硬化剤（E）	2,4,6-トリス(ジメチルアミノメチル)フェノール	5	5	5	2.5	2.5	2.5	2.5	5	5	5	5	5
		トリエチレンテトラミン				5	5	5	5					
	硬化触媒	AC *22				1								
		J1					1							
		J2						1						
		J3 *23	1	1	1					1	1	1	1	1
		DBTDL *24				0.2								
		J4 *30							1					
試験結果		取り扱い性	○	○	○	○	○	○	○	○	×	○	○	○

*1 ビスフェノールA型エポキシ樹脂（油化シェルエポキシ）
*3 膠質炭酸カルシウム（竹原化学工業（株））
*4 重質炭酸カルシウム（白石カルシウム工業（株））
*5 CEL-STAR SX-39（東海工業）
*6 ユーカーフェノリックマイクロバルーンズ BJO-0930（ユニオンカーバイド）
*7 マツモトマイクロスフェアーMFL-100CA（松本油脂製薬）
*8 マツモトマイクロスフェアーMFL-80GCA（松本油脂製薬）
*9 酸化チタン（石原産業）
*10 炭化水素系高沸点溶剤（日本石油化学）
*12 3－アミノプロピルトリメトキシシラン
*16 ビニルトリメトキシシラン
*17 テトラエトキシシラン
*18 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*19 ヒンダードアミン系光安定剤（旭電化工業株式会社）
*20 ベンゾトリアゾール系光安定剤（チバ・スペシャルティ・ケミカルズ株式会社）
*21 ヒンダードフェノール系酸化防止剤（チバ・スペシャルティ・ケミカルズ株式会社）
*22 ジブチルスズビスアセチルアセトナート
*23 ジブチルスズオキシドと酢酸との反応物
*24 ジブチルスズジラウレート
*25 フタル酸ジ2－エチルヘキシル
*26 水添ひまし油（楠本化成）
*27 フェニルオキシトリメトキシシラン
*28 ヘキサメチルジシラザン
*29 骨格アクリレート（東亜合成化学工業）
*30 3－エチルヘキサンスズ／ラウリルアミン＝3／1の混合・反応物

[0091]The following examinations were done about the constituent (11 points) which was good among the obtained constituents as for handling nature. A result is shown in Table 2-2.
(Tuck free examination) The tuck free examination was done based on JIS A5758. Uniformly, after mixing, on the glass plate, the spatula was used for about 3 mm in thickness, and base resin and a hardening agent were sounded level, and were placed promptly at the normal condition. The time which it took that a constituent atops adhering to a fingertip since the surface waa lightly touched and accustomed it by the fingertip purified by ethanol level was measured, and it was considered as tack free time.
(Tension ahear test) In [mix A agent and B agent uniformly, and] the bottom of 50% of humidity at

23 **, To an aluminum plate (25 mm in width, 100 mm in length, and 3 mm in thickness), an adhesion area of 25 mm x 25 mm, It applied so that it might be set to about 0.2 mm in thickness, and in 30 seconds, one more piece of aluminum plate is piled up alternately, and was stuck, and tensile shear bond strength (unit: kg/cm^2) was measured after care of health for seven days on the conditions with 5-mm the speed of testing for /.
[0092]
[Table 4]

表2－1

		組成物番号	実施例 D13	D14	D15	D16	D17	D18	D19	D20	比較例 D22	D23	D24
配合（A剤）	エポキシ樹脂（C）	エピコート828 *1	25	25	25	25	25	25	25	50	25	25	
	充填剤	ネオライトSP-T *3	20	20	20	20	20	20	20	20	20	20	
		ホワイトンSB *4	45	45	45	45	45	45	45	45	45	45	
	顔料	KR380 *9	5	5	5	5	5	5		5	5	5	
	可塑剤	ハイゾール *10	5	5	5	5	5	5	5		5	5	
（B剤）	ポリプロピレンオキシド	P1	56.3	51.5	18.8	37.5				25	75		50
		P2					37.5	37.5					
		P3							37.5				
		P4	18.8	31.5	56.3		37.5		37.5	25		75	50
		P5				37.5		37.5					
		P6											
	充填剤	ネオライトSP-T *3	30	30	30	30	30	30	30	30	30	30	50
		ホワイトンSB *4	60	60	60	60	60	60	60	60	60	60	100
		中空体1 *5				1							
		中空体2 *6					1						
		中空体3 *7						1					
		中空体4 *8							1				
	可塑剤	ハイゾール *10	5	5	5					5	5	5	10
		G1					20						
		G2						20					
		DOP *25				5							
	チキソ性付与剤	ディスパロン6500 *26	3	3	3					3	3	3	3
	シラン化合物（F）	アミノシラン1 *11	1	1	1	1	1	1	1	1	1	1	1
	貯蔵安定剤	VTMS *16	2	2	2					2	2	2	2
		TES *17				2	2	2	2				
	老化防止剤	アデカスタブLA62 *18					1		0.5				
		アデカスタブLA63P *19						1	0.5				
		チヌビン327 *20	1	1	1	1	1	1	1	1	1	1	1
		イルガノックス1010 *21	1	1	1	1	1	1	1	1	1	1	1
	モジュラス調整剤	L1					0.7		0.4				
		L2				1.3							
		L3 *27							0.4				
		L4 *28						0.5					
	空気硬化性化合物	桐油	5	5	5	5	5	5		5	5	5	5
	光硬化性化合物	アロニックスM309 *29	3	3	3		3	3	10	3	3	3	3
	エポキシ硬化剤（E）	2,4,6-トリス(ジメチルアミノメチル)フェノール	5	5	5	2.5	2.5	2.5	2.5	5	5	5	5
		トリエチレンテトラミン				5	5	5	5				
	硬化触媒	AC *22				1							
		J1					1						
		J2						1					
		J3 *23	1	1	1					1	1	1	1
		DBTDL *24				0.2							
		J4 *30							1				
試験結果		タックフリータイム[hr]	1.6	1.0	0.5	0.7	0.8	0.7	0.9	2.3	23	0.3	0.8
		引張剪断接着強さ[kg/cm²]	37	35	31	38	38	37	30	50	35	32	7

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*28 ヘキサメチルジシラザン
*30 特殊アクリレート（東亞合成化学工業）

[0093]
[Effect of the Invention]As explained above, according to this invention, the polymer which has a hydrolytic silicon group industrially obtained easily considering the silane compound [handling / a silane compound] as a raw material is used, It is dealt with by hypoviscosity, and excels in a sex, and

excels in the mechanical strength of the hardened material after hardening, and adhesive strength with adherend, and, moreover, the hardenability constituent which can be adjusted in the range with a wide cure rate can be obtained.

[Translation done.]